The Nexus between Reactive MD Simulations of RDX and the Reactive Euler Equations

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In the past we have focused on pre-ignition phenomena that cause a local temperature rise when a single void in high-melting explosive (HMX) collapses under the action of a shock wave. Working in the melting regime, a timeline can be associated with the collapse of a single void through a consideration of the time scales on which these mechanisms are activated.

Among the phenomena we studied, the hydrodynamic mechanism is generally considered to be an important pre-ignition step. In this mechanism, the shock-driven incident side of the void impinges on the shadow side of the void and is brought to rest, causing a considerable temperature rise in the HMX. This mechanism comes into consideration as the void closes up under the action of the shock wave. The resulting increase in pressure and temperature causes a shock wave to emanate from the collapse. This shock wave very quickly attains cylindrical symmetry in our two-dimensional (2D) calculation. (See Fig. 1.) If the energy contained in this outgoing shock wave is sufficiently high, and the size of the hotpsot is sufficiently large, it could produce a local microdetonation.

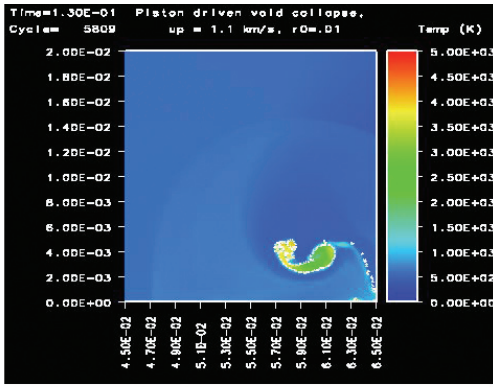

Figure 1—
Numerical simulation of a collapsing spherical void approximately 0.145 microseconds after the launch of a shock wave.

In order to study the energy release provided by the hydrodynamic collapse of the void, one needs to understand the chemical reaction pathway through which this homogeneous explosive decomposes. Very recently A. Strachan (T-14) found that the complicated reaction pathway in Royal Demolition Explosive (RDX) can be simply represented as a single-step energy release step governed by Arrhenius kinetics. The activation energy is about 1 eV and is weakly pressure-dependent. This result was obtained by using the ReaxFF potential developed at California Institute of Technology in cook-off-like simulations of RDX at high temperature and pressure. These simulations were performed at various temperatures and pressures, and it is of interest to investigate the applicability of the resulting parameterization of the kinetics at a macroscopic scale.

The incorporation of the single-step Arrhenius rate into continuum dynamics can be accomplished by solving the reactive Euler equations. The width of the reaction zone which develops as the rarefaction Taylor wave behind the von Neumann pressure spike may be estimated in an order of magnitude sense by the product of the sound speed (typically a kilometer per second or less) and the characteristic chemical time scale, viz., picoseconds. This yields a scale of the order of 1/100th of a micron. Since voids in explosives are expected to be of the order of microns, it follows that a numerical solution of the reactive Euler equations would be at best a difficult job in two dimensions.

An alternative solution is to develop a subgrid model, such as Bdzil's Detonation Shock Dynamics (DSD). This technique consists of performing an asymptotic (perturbative) expansion of the reactive Euler equations in a shock-based frame of reference and coordinate system. The formalism eventually reduces to the solution of a transcendental equation for the relation between the detonation velocity normal to the shock front (D_n) and the local curvature (κ). It is possible to obtain from this analysis an estimate of the minimum size and energy of the hotspot that would be required to sustain a microdetonation during void collapse in a melted homogeneous RDX sample.

The reaction rate obtained by Strachan can be written down as:

$$
\begin{aligned}
R(T,\rho) &= \kappa(\rho)\exp(-(E_a + f\rho_a/\rho)\kappa T)(pico - s)\\
\kappa(\rho) &= \exp(a - b\rho_a/\rho)\\
\underline{a} &= 1.82561\\
b &= 0.290392\\
f &= 0.356\\
E_a &= 21.472 \text{ (kcal/mol)}
\end{aligned}
\qquad (1)
$$

where ρ_a is the initial density of the material.

If T_{CJ} is the Chapman-Jouguet temperature, the rate R may be written as:

$$
\begin{aligned}
R(T,\rho) &= \kappa \exp(-1/\epsilon)\exp\left(\frac{T - T_{CJ}}{\epsilon}\right)\\
&\qquad \exp(-f\rho_a/\rho/\kappa T + E_a/\kappa T_{CJ})\\
\epsilon &= \frac{\kappa T_{CJ}}{E_a} \sim 0.13\\
T - T_{CJ} &\sim \epsilon \quad .
\end{aligned}
\qquad (2)
$$

Thus ϵ provides one with a small dimensionless parameter with which to perform an asymptotic analysis of the reactive Euler equations. One can now use the methods developed in [1], where the reactive Euler equations are written in the shock-based frame of reference to obtain a formal solution to $\vartheta(\epsilon)$ as:

- Note that an extra assumption has been made in this model, viz., that the factor of $(1 - \lambda_r)$ which appears in the conventional form for the single-step Arrhenius rate has been replaced by $(1 - \lambda_r)^{1/2}$. Physically this allows the reaction to be complete within a reaction zone of finite width. This is a reasonable assumption as long as the major fraction of the energy is released within a short distance. We have experimented with retaining the linear factor of $(1 - \lambda_r)$ in our analysis, unsuccessfully.
- Furthermore, for the moment we utilized a polytropic equation of state to describe the explosive. We expect to replace it shortly with a realistic equation of state obtained directly from the MD simulations.

A nonlinear $D_n - \kappa$ relation was obtained from this formalism and used to compute the shape of a steady-state detonation front for the case of an unconfined cylinder of macroscopic dimension (Fig. 2), thereby completing the initial phase of the transportation of the reactive MD simulations to the continuum level.

[1] M. Short and J.B. Bdzil, "Propagation Laws for Steady Curved Detonations with Chain-Branching Mechanisms," *J. Fluid Mech.* **479**, 39-64 (2003).

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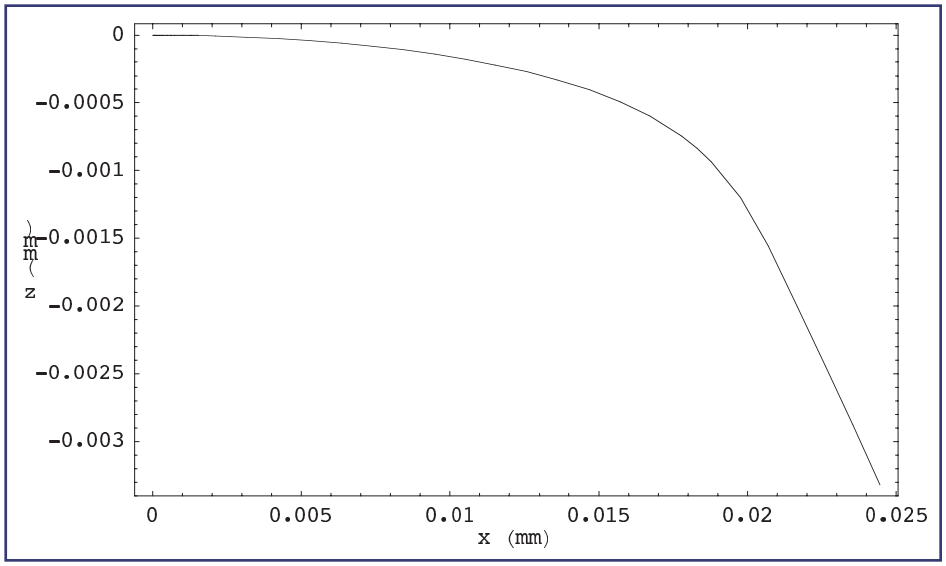

Figure 2—
Shape of the detonation front for an unconfined cylinder.